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Growth factors for Marangoni instability in a spherical liquid layer under zero-gravity conditions

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Abstract. The neutral-stability analysis presented by Hoefsloot et al. [3] is completed by computing the growth factors β for the normal modes and by showing that the neutral states ($\mathrm{Re}(\beta)=0$) are stationary ($\mathrm{Im}(\beta)=0$) rather than oscillatory ($\mathrm{Im}(\beta) \neq 0$).

1. Introduction

Hoefsloot et al. [3] studied the onset of surface-tension-driven convective flows (Marangoni instability) for a spherical liquid layer under zero-gravity conditions. The layer contains a solute that can evaporate at the inner surface (a spherical gas/liquid interface) and the outer sphere is taken to be a rigid solid surface. Initially the solute concentration is uniformly equal to c_0. Two separate cases are distinguished: the outer surface is impervious (case 1) and the solute concentration has the constant value c_0 at the outer surface (case 2). By use of a linear normal-mode analysis neutral (or marginal) stability conditions have been computed in [3] for concentration profiles created by diffusion in the motionless liquid, which profiles were 'frozen' at a certain point in time. Neutral stability was investigated by putting the growth factor (β) equal to zero. This leads to an eigenvalue problem yielding for each mode the neutral-stability value of the Marangoni number Ma. The so-called critical Marangoni number (Ma_c) is equal to the smallest of the Ma-values belonging to all possible modes; the corresponding wavenumber (α_c) is called the critical wavenumber. If Ma exceeds slightly the neutral-stability value of a particular mode, this mode will grow in time, the initial rate of growth being determined by β. The principal purpose of the present paper is to compute the growth factors β for the stability problems treated in [3].

In the analysis of [3], and also in the related paper [2], it has been assumed that the so-called 'principle of exchange of stability' holds, that is, if $\mathrm{Re}(\beta)=0$ then automatically $\mathrm{Im}(\beta)=0$. This means that the neutral states are stationary rather than oscillatory ($\mathrm{Im}(\beta) \neq 0$). Vidal and Acrivos [4] have shown that this principle is valid for certain systems with a *flat* interface in which surface-tension effects are solely

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responsible for instability. The only cases for which oscillatory neutral states have been found, appear to possess two competing instability mechanisms. It is also the purpose of this paper to show, by numerical means, that no oscillatory neutral modes exist for the spherical systems under consideration.

2. The eigenvalue problem for the growth factors β

First, we recapitulate the mathematical formulation of the stability problem [3]. Throughout the present paper we adhere to the definitions, notations and non-dimensionalization introduced in [3] and for a discussion of the assumptions underlying the mathematical model we also refer to that paper. For the spherical liquid layer (inner radius a, layer thickness H), the two non-dimensional equations in spherical coordinates (r, θ, φ) governing the perturbation of a given motionless basic concentration profile $c_b(r)$ are [3]:

$$\nabla^2\left(\mathrm{Pr}^{-1}\frac{\partial}{\partial t} - \nabla^2\right)(ru) = 0, \tag{2.1}$$

$$\left(\frac{\partial}{\partial t} - \nabla^2\right)c = -u\,\frac{\mathrm{d}c_b}{\mathrm{d}r}, \tag{2.2}$$

where

$$\nabla^2 = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right) - \frac{1}{r^2}\mathfrak{L}^2, \quad \mathfrak{L}^2 = \frac{-1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) - \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}.$$

In these equations, u denotes the radial liquid velocity, c is the perturbation solute concentration, t is the time variable, and Pr is the Prandtl number. The boundary conditions at the gas/liquid interface $r = a/H$ are

$$u = 0, \qquad \frac{\partial c}{\partial r} = \mathrm{Bi}\,c, \qquad \frac{\partial^2(ru)}{\partial r^2} = \frac{1}{r}\mathrm{Ma}\,\mathfrak{L}^2 c, \tag{2.3}$$

where Bi and Ma denote the Biot and Marangoni number, respectively. At the solid boundary $r = 1 + a/H$ we have the conditions

$$u = \frac{\partial u}{\partial r} = 0, \qquad \frac{\partial c}{\partial r} = 0 \text{ (case 1)} \quad \text{or} \quad c = 0 \text{ (case 2)}. \tag{2.4}$$

The solution of the boundary-value problem (2.1)–(2.4) is now written in separated form (normal modes):

$$ru(r,\theta,\varphi,t)=(U(r)+i\mu(r))Y_n^m(\theta,\varphi)\,e^{\beta t},$$
$$c(r,\theta,\varphi,t)=(C(r)+i\gamma(r))Y_n^m(\theta,\varphi)\,e^{\beta t},\quad m,n=1,2,3,\ldots, \tag{2.5}$$

where $Y_n^m(\theta,\varphi)$ are spherical surface harmonics of the first kind [1], $U+i\mu$ and $C+i\gamma$ denote the complex amplitude of the radial velocity and concentration perturbation, and $\beta=p+iq$ is the complex growth factor of the mode (m,n). Instability is characterized by $p>0$, neutral stability by $p=0$ and stability by $p<0$. If $q\neq 0$ for $p=0$, we have an oscillatory neutral state.

After substitution of (2.5) in problems (2.2)–(2.4) and subsequently separating real and imaginary parts, we obtain four ordinary differential equations for $U(r)$, $\mu(r)$, $C(r)$ and $\gamma(r)$,

$$(\mathfrak{D}-\mathrm{Pr}^{-1}p)\mathfrak{D}U(r)+\mathrm{Pr}^{-1}q\mathfrak{D}\mu(r)=0, \tag{2.6}$$

$$(\mathfrak{D}-\mathrm{Pr}^{-1}p)\mathfrak{D}\mu(r)-\mathrm{Pr}^{-1}q\mathfrak{D}U(r)=0, \tag{2.7}$$

$$(\mathfrak{D}-p)C(r)+q\gamma(r)=\frac{U(r)}{r}\frac{\mathrm{d}c_b}{\mathrm{d}r}, \tag{2.8}$$

$$(\mathfrak{D}-p)\gamma(r)-qC(r)=\frac{\mu(r)}{r}\frac{\mathrm{d}c_b}{\mathrm{d}r}, \tag{2.9}$$

with

$$\mathfrak{D}=\frac{\mathrm{d}^2}{\mathrm{d}r^2}+\frac{2}{r}\frac{\mathrm{d}}{\mathrm{d}r}-\frac{n(n+1)}{r^2}$$

and boundary conditions at $r=a/H$:

$$U=\mu=0,\qquad \frac{\mathrm{d}C}{\mathrm{d}r}=\mathrm{Bi}\,C,\qquad \frac{\mathrm{d}\gamma}{\mathrm{d}r}=\mathrm{Bi}\,\gamma,$$
$$\frac{\mathrm{d}^2U}{\mathrm{d}r^2}=\mathrm{Ma}\,\frac{n(n+1)}{r}C,\qquad \frac{\mathrm{d}^2\mu}{\mathrm{d}r^2}=\mathrm{Ma}\,\frac{n(n+1)}{r}\gamma \tag{2.10}$$

and at $r=1+a/H$:

$$U=\frac{\mathrm{d}U}{\mathrm{d}r}=0,\qquad \mu=\frac{\mathrm{d}\mu}{\mathrm{d}r}=0, \tag{2.11}$$

$$\frac{\mathrm{d}C}{\mathrm{d}r}=\frac{\mathrm{d}\gamma}{\mathrm{d}r}=0\ \text{(case 1)}\quad\text{or}\quad C=\gamma=0\ \text{(case 2)}. \tag{2.12}$$

Notice that m, the wavenumber in the φ-direction, has vanished from the problem; only the θ-wavenumber n plays a role.

Boundary-value problem (2.6)–(2.12), with $c_b(r)$, a/H, n, Bi, Pr and Ma given, is a linear eigenvalue problem for the eigenvalues $\beta = p + iq$ and the eigensolutions $U + i\mu$ and $C + i\gamma$. We shall assume that there exists an infinite denumerable sequence of eigenvalues β_j which can be ordered such that $\mathrm{Re}(\beta_{j+1}) < \mathrm{Re}(\beta_j)$, $j = 0, 1, 2, \ldots$. This assumption turns out to be supported by our numerical results.

3. Numerical solutions of the eigenvalue problem

The eigenvalue problem consisting of equations (2.6)–(2.9) and boundary conditions (2.10)–(2.12) has been solved numerically with a shooting technique. First, the basic concentration profile $c_b(r)$ is computed for given values of a/H and Bi and the desired time t at which the diffusion process in the motionless liquid is frozen. Details of this computation can be found in [3]. Next, for given values of n and Ma and an *estimated* value of the growth factor $\beta = p + iq$, the general solution of the following pair of initial-value problems A and B is determined:

—problem A for (U, μ) consists of equations (2.6), (2.7) and initial conditions (2.11) at the solid boundary $r = 1 + a/H$;
—problem B for (C, γ) consists of equations (2.8), (2.9) together with initial conditions (2.12) at $r = 1 + a/H$.

The general solution of problem A can be written as

$$(U, \mu) = \sum_{j=1}^{4} B_j(U_j, \mu_j), \tag{3.1}$$

where $B_1, \ldots, B_4$ are integration constants and U_j and μ_j satisfy the additional initial conditions

$$(U_j'', U_j''', \mu_j'', \mu_j''') = \mathbf{e}_j \quad (j = 1, \ldots, 4) \text{ at } r = 1 + a/H$$

where $\mathbf{e}_j$ denotes the jth unit vector in $\mathbb{R}^4$ (or, in other words, the jth row of the 4×4 unit matrix). Likewise, the general solution of problem B is given by

$$(C, \gamma) = \sum_{j=1}^{4} B_j(C_j, \gamma_j) + B_5(C_5, \gamma_5) + B_6(C_6, \gamma_6), \tag{3.2}$$

where the summation term represents a particular solution of (2.8)–(2.9) in which (C_j, γ_j), $j = 1, \ldots, 4$, satisfies the initial conditions at $r = 1 + a/H$:

$$(C_j, C_j', \gamma_j, \gamma_j') = (0, 0, 0, 0).$$

The last two terms at the right of (3.2) involve the integration constants B_5 and B_6 accompanied by the two homogeneous solutions (C_j, γ_j), $j = 5, 6$, of equations (2.8), (2.9) satisfying the initial conditions at $r = 1 + a/H$:

$$(C_j, C_j', \gamma_j, \gamma_j') = \begin{cases} (1, 0, 0, 0) \text{ for } j = 5 \\ (0, 0, 1, 0) \text{ for } j = 6 \end{cases} \quad \text{(case 1)},$$

or

$$(C_j, C_j', \gamma_j, \gamma_j') = \begin{cases} (0, 1, 0, 0) \text{ for } j = 5 \\ (0, 0, 0, 1) \text{ for } j = 6 \end{cases} \quad \text{(case 2)}.$$

The fundamental solutions (U_j, μ_j), $j = 1, \ldots, 4$ and (C_j, γ_j), $j = 1, \ldots, 6$, have been computed with a fourth-order Runge–Kutta method.

Substituting the combined general solution (3.1), (3.2) of problems A and B in the remaining six boundary conditions (2.10) at the gas/liquid interface $r = a/H$, we obtain a set of six homogeneous linear algebraic equations for the integration constants $B_1, \ldots, B_6$. The coefficient determinant D of this set of equations depends on the value of β. If $D(\beta) = 0$, then β is an eigenvalue; if not, then the estimate for β has to be improved successively until $D(\beta)$ becomes sufficiently small.

Numerical computation of $D(\beta)$ for a number of cases shows that $D(\beta)$ will not vanish unless $\mathrm{Im}(\beta) = q = 0$ (see next section). So, in the subsequent computation of the growth factors β and eigensolutions (U, C), it is allowed to take β real $(q = 0)$ and, without loss of generality, to consider only real eigensolutions, that is, to put $\gamma(r) = \mu(r) = 0$. Accordingly, the shooting method described above simplifies considerably. The combined general solution of problems A and B now consists of five fundamental solutions accompanied by only three integration constants (analogous to the shooting method used in [3]):

$$\begin{aligned} U(r) &= B_1 U_1(r) + B_2 U_2(r), \\ C(r) &= B_1 C_1(r) + B_2 C_2(r) + B_5 C_5(r). \end{aligned} \tag{3.3}$$

The eigenvalue equation for β follows from the equating to zero of a 3×3 coefficient determinant and the zeros of this equation have been computed by use of the Regula Falsi method.

The eigenvalue problem turns out to possess a denumerably infinite number of eigenvalues. We are mainly interested in the largest eigenvalue, β_0, because this value determines the stability behaviour of the particular mode under consideration. In order to decide whether a computed eigenvalue is indeed the largest one, the corresponding eigensolution $(U(r), C(r))$ is computed; U and C are found as particular linear combinations of the form (3.3). Typically, the eigensolution belonging to the largest eigenvalue β_0 possesses no zeros in the *interior* of the interval $a/H \leqslant$

$r \leqslant 1 + a/H$. So the search for zeros of $D(\beta)$ is continued until the one is found for which $(U(r), C(r))$ has no zeros in the open interval $(a/H, 1 + a/H)$.

4. Numerical results and discussion

The possible occurrence of oscillatory neutral states has been investigated by performing a series of computations in which the coefficient determinant $D(\beta)$ has been determined under the assumption that β is purely imaginary ($\beta = iq$, $p = 0$). Figure 1 is a typical result for case 1. It shows D as a function of q for various values of Ma (and fixed parameter values Pr = 787, $a/H = 10$, Bi = 20, $t = 0.001$) for the mode with wavenumber $\alpha = 9$ ($\alpha = nH/a$ is the number of waves in the θ-direction per unit length along the interface $r = a/H$). This mode is the critical one with regard to neutral stability for this particular set of parameter values [3] with corresponding $\mathrm{Ma}_c = 1058$. It is seen that D is an even function of q, as can be shown easily by observing that, if $(U, \mu, C, \gamma, p, q)$ satisfies boundary-value problem (2.6)–(2.12), then also $(\mu, U, \gamma, C, p, -q)$ is a solution. Figure 1 shows that D does not vanish unless Ma has its critical value and, furthermore, if D vanishes, then $q = 0$. Figure 2 presents analogous results for case 2. The general pattern of Figs 1 and 2 has also been found for a large number of other sets of parameter values. On the basis of these findings it can be concluded that there are no oscillatory neutral states and, hence, that the principle of exchange of stability holds for the spherical systems under consideration.

The question of the possibility of oscillatory growth ($p > 0, q \neq 0$) has also been addressed. Figures 3 and 4 show typical results for case 1 and case 2, respectively. Here D has been plotted as a function of p for several values of q for fixed values of Pr, a/H,

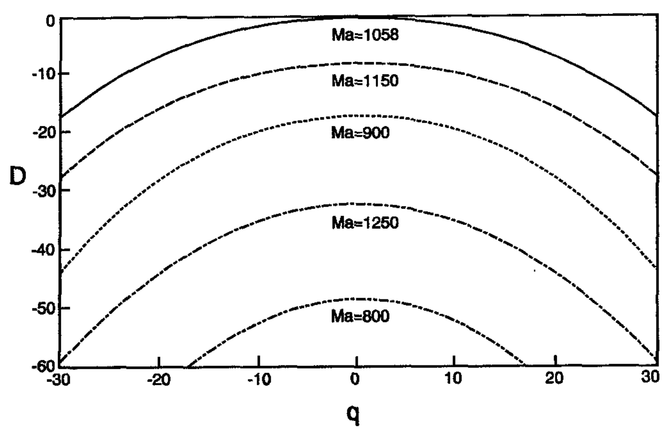

Fig. 1. Determinant D for case 1 as a function of $q(=\mathrm{Im}(\beta))$ for various values of Marangoni number Ma. Parameter values: $\mathrm{Pr} = 787$, $a/H = 10$, $\alpha = 9$, $\mathrm{Bi} = 20$, $t = 0.001$, $p(=\mathrm{Re}(\beta)) = 0$. The corresponding critical values are $\mathrm{Ma}_c = 1058$ and $\alpha_c = 9$.

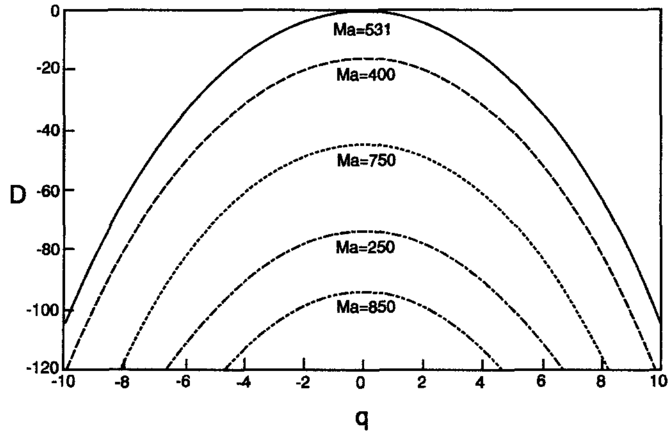

Fig. 2. Determinant D for case 2 as a function of $q(=\text{Im}(\beta))$ for various values of Marangoni number Ma. Parameter values: $\text{Pr}=1$, $a/H=1$, $\alpha=4$, $\text{Bi}=20$, $t=0.1$, $p(=\text{Re}(\beta))=0$. Here, $\text{Ma}_c=531$ and $\alpha_c=4$.

α, Bi and t. The Marangoni number has been chosen as $\text{Ma}=1200$, which is slightly above the critical value for both cases and it leads to at least one mode growing in time. In both figures it is seen that D does not vanish for $p>0$ unless $q=0$ with corresponding real growth factor $p=\beta_0>0$. So the growth of the modes under unstable circumstances is governed by a real exponential function of time, rather than a complex exponential, the latter of which would imply oscillatory growth in time with exponentially increasing amplitude.

The numerical results discussed so far show that in the subsequent computations, the results of which will be presented below, we can restrict ourselves to the case of

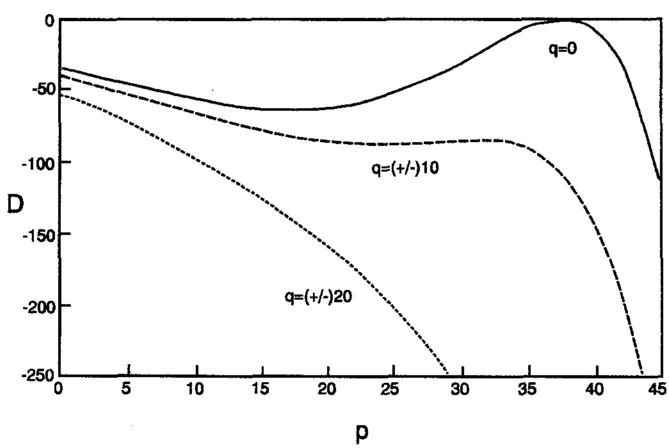

Fig. 3. Determinant D for case 1 as a function of $p(=\text{Re}(\beta))$ for various values of $q(=\text{Im}(\beta))$. Parameter values: $\text{Pr}=787$, $a/H=10$, $\alpha=9$, $\text{Bi}=20$, $\text{Ma}=1200$, $t=0.001$. ($\text{Ma}_c=1058$ and $\alpha_c=9$.)

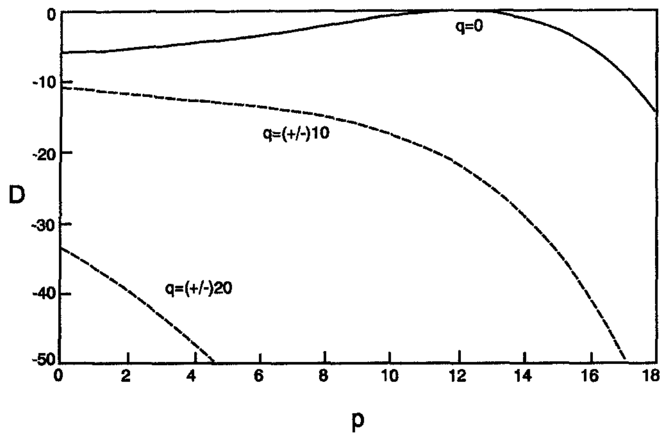

Fig. 4. Determinant D for case 2 as a function of $p(=\mathrm{Re}(\beta))$ for various values of $q(=\mathrm{Im}(\beta))$. Parameter values: $\mathrm{Pr}=1$, $a/H=1$, $\alpha=4$, $\mathrm{Bi}=40$, $\mathrm{Ma}=1200$, $t=1.0$ ($\mathrm{Ma}_c=1115$ and $\alpha_c=4$.)

real growth factors β with corresponding real eigensolutions pairs $(U(r), C(r))$. As explained in Section 3, this means a considerable simplification of the numerical shooting method.

Figures 5 and 6 show the growth factor β_0 for cases 1 and 2, respectively, as a function of the wavenumber α for several values of the 'freezing' time t at which the

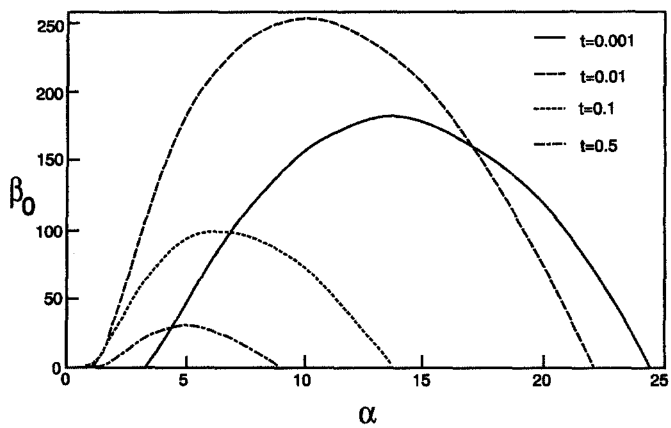

Fig. 5. Growth factor β_0 for case 1 as a function of wavenumber α for various values of time t. Parameter values: $\mathrm{Pr}=787$, $a/H=1$, $\mathrm{Bi}=20$, $\mathrm{Ma}=1600$. The critical values are as follows:

t	0.001	0.01	0.1	0.5
Ma_c	1071	505	516	830
α_c	9	5	4	3

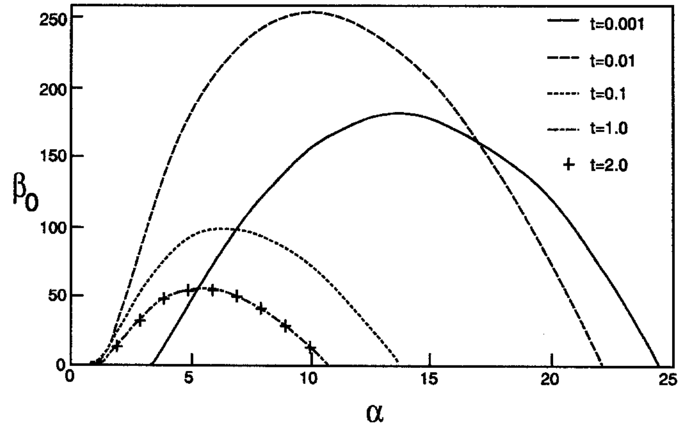

Fig. 6. Growth factor β_0 for case 2 as a function of wavenumber α for various values of time t. Parameter values: $\mathrm{Pr}=787$, $a/H=1$, $\mathrm{Bi}=20$, $\mathrm{Ma}=1600$. The critical values are as follows:

t	0.001	0.01	0.1	1.0	2.0
Ma_c	1071	508	531	649	649
α_c	9	5	4	4	4

basic motionless concentration profile $c_b(r)$ has been obtained. In both cases the chosen Marangoni number ($\mathrm{Ma}=1600$) is larger than the values of Ma_c belonging to the various times involved, so there is always growth ($\beta_0 > 0$) for some interval of α-values at each time instant for which the graphs have been presented. It is seen that the maximum growth factor increases initially and decreases for later times. In case 1 there will be no positive values of β_0 after some finite time, when Ma_c has become larger than 1600. In case 2 the graphs tend to a limit graph which has been reached almost completely for $t=1$; this corresponds to the non-zero limit concentration profile being reached for $t \to \infty$ in this case. For small times the graphs for cases 1 and 2 are nearly identical, since for these times the difference in the boundary condition at the solid outer surface is not yet noticeable in the basic concentration profile $c_b(r)$. The wavenumber α at which β_0 reaches its maximum value decreases in time. This is due to the increasing penetration depth of the motionless diffusion process as time increases, allowing for larger wavelengths of the normal modes. These α-values are always larger than the corresponding values of α_c at each time.

Table 1 shows the strong dependence of β_0 on the curvature parameter a/H and Biot number Bi for fixed time $t=0.01$, wavenumber $\alpha=5$, Prandtl number $\mathrm{Pr}=787$ and various values of Ma. For fixed Bi and Ma the largest curvature ($a/H=0.1$) shows the lowest value of β_0. Increasing the value of a/H from 1 to 10 leads in most cases to an increase of β_0 for $\mathrm{Bi}=1$ and 20, and to a decrease of β_0 for $\mathrm{Bi}=40$ and $\mathrm{Bi}=60$.

Table 1. Results for case 1: growth factor β_0 as a function of a/H for Bi = 1, 20, 40, 60 and various values of Ma. Parameter values: Pr = 787, $\alpha = 5$, $t = 0.01$

	Bi = 1			Bi = 20		
a/H	Ma = 1100	Ma = 2000	Ma = 3000	Ma = 600	Ma = 2000	Ma = 3000
0.1	−1.4	−1.4	−1.2	−1.3	53.0	160.7
1	1.8	37.8	80.5	12.3	254.8	430.0
10	9.0	50.1	96.0	14.8	254.1	490.3

	Bi = 40			Bi = 60		
a/H	Ma = 800	Ma = 1400	Ma = 2000	Ma = 1100	Ma = 1500	Ma = 2000
0.1	−1.3	0.3	27.9	−1.3	−0.7	6.1
1	3.9	76.3	160.8	5.3	40.6	92.9
10	3.3	75.3	155.9	3.7	38.7	88.6

Table 2. Values of β_0, β_1, β_2 and β_3 for case 1 for various values of Pr. Parameter values: $a/H = 10$, $\alpha = 9$, Bi = 20, Ma = 1200, $t = 0.001$ (Ma$_c$ = 1058, $\alpha_c = 9$)

Pr	β_0	β_1	β_2	β_3
0.1	3.1	−9.4	−14.7	−22.3
1	16.3	−73.7	−91.9	−99.8
787	34.9	−73.8	−101.1	−149.2

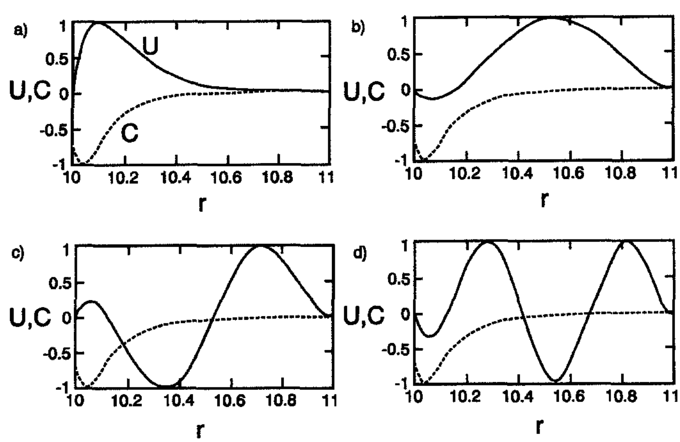

Fig. 7. Eigensolutions U and C for case 1 as a function of the radial coordinate r, corresponding to the eigenvalues β_0, β_1, β_2 and β_3. Parameter values: Pr = 0.1, $a/H = 10$, $\alpha = 9$, Bi = 20, Ma = 1200, $t = 0.001$ (Ma$_c$ = 1058, $\alpha_c = 9$). Figure (a): (U_0, C_0), (b): (U_1, C_1), (c): (U_2, C_2), (d): (U_3, C_3).

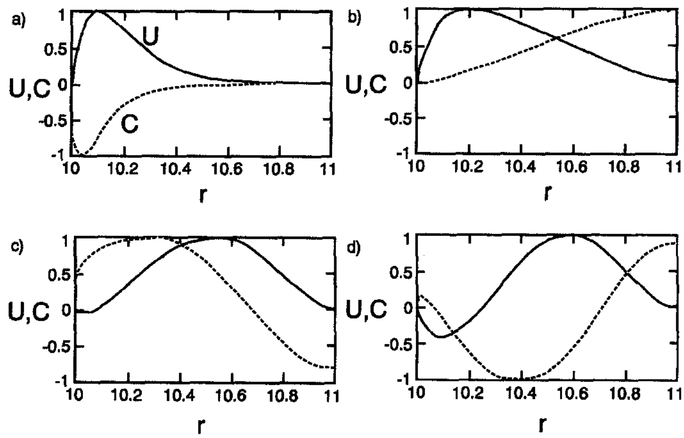

Fig. 8. As for Fig. 7 with Pr = 1.

The dependence of the first four eigenvalues β_0, β_1, β_2 and β_3 on the Prandtl number Pr for fixed values of a/H, α, Bi, Ma and t is shown in Table 2 for case 1. Increasing values of Pr are seen to yield increasing values of $|\beta_i|$, $i = 0, 1, 2, 3$.

For the same parameter values as in Table 2, Figures 7, 8 and 9 show for case 1 the dependence of the first four eigensolution pairs (U_i, C_i), $i = 0, 1, 2, 3$, on Pr. It is remarkable that for Pr small (large) the eigensolution $C_i(U_i)$ has about the same shape for $i = 0, 1, 2, 3$. A nice property of the computed eigensolution pairs is seen to be that U_i and C_i together possess precisely i zeros in the open interval $a/H < r < 1 + a/H$.

The increase of the penetration depth of the diffusion process as time progresses is clearly demonstrated by the graphs of U_0 and C_0 for various times t for case 1 (see Fig. 10).

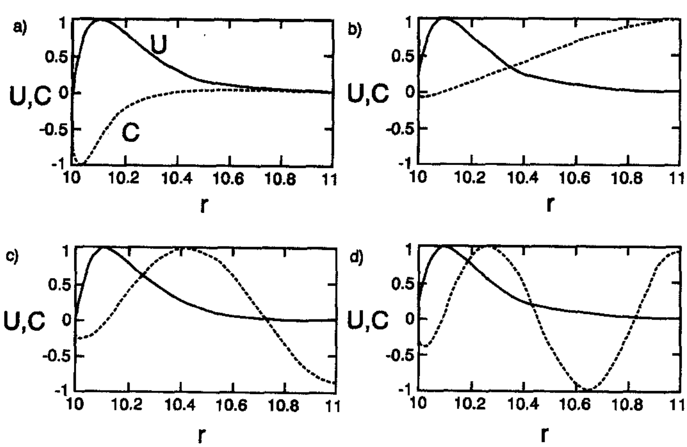

Fig. 9. As for Fig. 7 with Pr = 787.

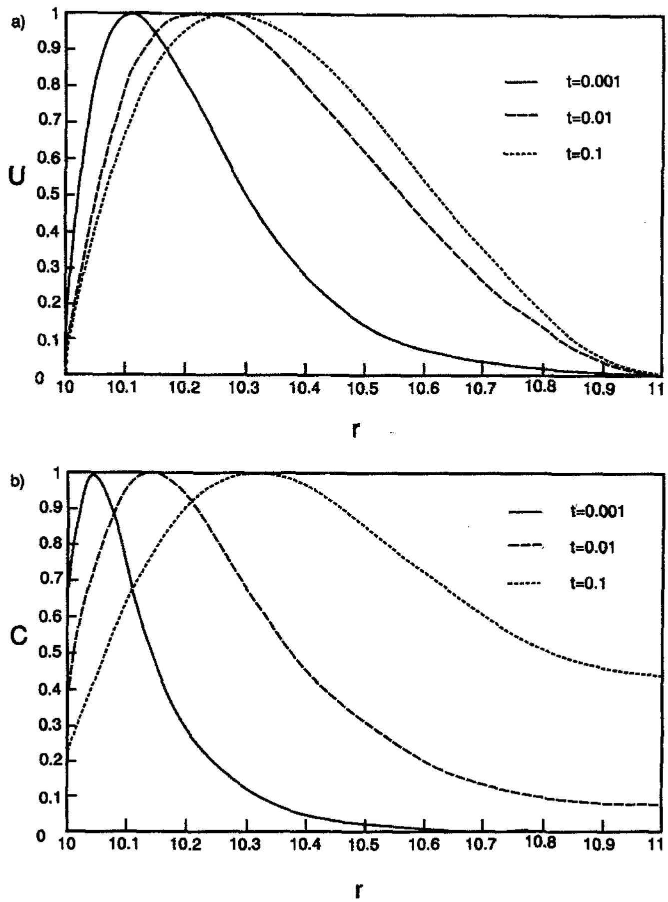

Fig. 10. Eigenfunctions U_0 (Fig. 10(a)) and C_0 (Fig. 10(b)) for case 1 as functions of r for $t=0.001$, 0.01 and 0.1. Parameter values: Pr = 787, $a/H=10$, Bi = 20. Ma and α (and corresponding value of the growth factor β_0) are as indicated in the table below:

t	Ma	α	β_0
0.001	1070	9.0	2.9
0.01	500	4.3	1.1
0.1	540	3.0	0.5

5. Concluding remarks

For the linear stability problem governing the onset of Marangoni convection in a spherical liquid layer under zero-gravity conditions, it has been shown that no oscillatory neutral modes exist. The only possible neutral modes are stationary.

Further, the initial growth in time of the modes under unstable circumstances is shown to be monotone rather than oscillatory with increasing amplitude.

The maximum of the growth factor, taken over all possible modes at a certain 'freezing' time t, is an increasing function of t for sufficiently small t and it decreases again for later times. Apart from the obvious dependence of the growth factor for a particular mode on the Marangoni number Ma, this growth factor also turns out to depend quite strongly on the Prandtl number Pr, the Biot number Bi and the curvature parameter a/H.

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